# Ultrafast Phase Transitions

*Shedding Light on the Unique Properties of "Correlated" Materials*

An LBNL team led by Robert Schoenlein and Andrea Cavalleri has measured the fundamental time scale for the switching of vanadium dioxide from an insulating to a conducting state. They showed that the transition occurs in 80 femtoseconds ($8x10^{-14}$ seconds), and from the data, have deduced critical clues to understanding the fundamental physics of this novel material.

Vanadium dioxide ($VO_2$) is a prototypical "correlated" material, in which the interactions among the electrons play a significant role in determining the material properties. Deciphering the electron interactions is a fundamental challenge in understanding a variety of these materials, including high-$T_c$ superconductors, and colossal magneto-resistance materials.

A unique property of $VO_2$ is that at very modest temperatures (above 67 °C) it transforms from an insulating to a metallic phase, with a 100,000-fold change in conductivity; it undergoes dramatic changes in reflectivity; and it also changes in crystal structure from "monoclinic" to "rutile". [The reflectivity properties are already being exploited for energy conserving, temperature-switchable (thermochromic) window coatings.] Although $VO_2$ is a relatively simple correlated material, and has been studied for decades, the driving force behind its remarkable changes in properties had not been elucidated. In particular, it was not known if changes in the crystal structure were primarily responsible for the changes in the electronic properties, or, alternatively, if the electronic properties (electron-electron correlation forces) drove the changes in the crystal structure.

The key to deciphering this phenomenon lay in measuring the time-dependent changes in the properties of $VO_2$ using femtosecond pulses from a state-of-the-art laser system. These measurements take advantage of the fact that the insulator-to-metal transition in $VO_2$ can be induced by light ("pump") pulse and can be observed by measuring the change in the material's reflectivity with a subsequent "probe" pulse. A series of such measurements, using progressively shorter intervals between the pump and probe pulses, shows that transformation to the metallic state takes a minimum of 80 femtoseconds. This time transition limit was identified as a "structural bottleneck" because it corresponds to the time required for the vanadium atoms to move from their positions in the insulating crystal structure to their positions in the metallic crystal structure. Additional measurements of the vibrational motion of the vanadium atoms showed that 80 femtoseconds corresponds to roughly half of a vibrational period. This demonstrates for the first time, that the insulator-to-metal transition in this material is primarily mediated by the motion of the atoms, and not by correlation among the electrons. The measurements further suggest that a simple coherent motion of the vanadium atoms is responsible for the transition.

In one of the first applications of ultrafast x-ray spectroscopy using a femtosecond beamline at the Advanced Light Source, the team has directly probed the ultrafast changes in the electronic density of states during this $VO_2$ transition. In these studies, the tunability of the femtosecond x-ray source is critical since it provides a means to selectively probe electronic energy levels of one symmetry from the vanadium absorption edge, and energy levels of a different symmetry from the oxygen absorption edge. Early results at two wavelengths indicate the collapse of the band-gap (which is indicative of the insulator to metal transition) on a sub-picosecond time scale. A more complete x-ray spectroscopy study of these changes in the electronic energy structure will become possible with a new femtosecond x-ray beamline that is now under construction. In the future, this same experimental approach may be applied to other correlated systems to understand the fundamental origins of their unique material properties.

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